

Supporting information

3D plasmonic chiral colloids

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A. Experimental Methods

1. Materials

HAuCl₄•3H₂O, Hexadecyltrimethyl ammonium bromide (CTAB), Sodium dodecyl sulfate (SDS), Tris (2-carboxyethyl) phosphine hydrochloride (TCEP), Bis (*p*-sulfonatophenyl) phenyl phosphine dihydrate dipotassium salt (BSPP), Silver nitrate (AgNO₃), Sodium borohydride (NaBH₄), L-ascorbic acid, Trisodium citrate dehydrate (C), and Citric acid were purchased from Sigma-Aldrich. All chemicals were used as received without further purifications. All oligonucleotides were purchased from Invitrogen.

2. Preparation of the DNA origami

Rectangular origami templates were prepared according to reference¹ with several changes. All of the side staples were left out to prevent aggregation via helix stacking interactions between adjacent origami. Three groups of different capture strands were designed to assemble AuNRs on the origami templates. These capture strands were extended by 15 bases with specific sequences. The rectangular DNA origami was annealed at a ratio of 1:5:10 for the M13 strand, capture strands, and staples strands. The DNA origami was subsequently assembled in a 1×TAE/Mg²⁺ buffer (Tris, 40 mM; Acetic acid, 20 mM; EDTA, 2 mM; and Magnesium acetate, 12.5 mM; pH 8.0) by cooling slowly from 90 °C to room temperature. The DNA origami was then filtered with 100 kDa (MWCO) centrifuge filters to remove the extra capture strands and staple strands.

3. Synthesis of the AuNRs

A two-step method was used to synthesize AuNRs according to reference².

a. Synthesis of the AuNR seeds

50 µL of 2% (w/v) HAuCl₄ solution and 7.5 mL of 100 mM CTAB solution were added to a 25 mL flask. A 600 µL of ice-cold NaBH₄ solution (10 mM) was added to the flask under vigorous

stirring. The color of the solution changed quickly to yellowish brown. The resulting solution acted as nucleation seeds for the AuNR synthesis in the next step.

b. Synthesis of the AuNRs

An 80 μL of 2% (w/v) HAuCl_4 solution was added to a 10 mL of 100 mM CTAB solution. 75 μL of 10 mM AgNO_3 solution and 50 μL of 100 mM L-ascorbic acid solution were added to the flask in order. A 20 μL seed solution was added to the mixture solution under stirring. The final solution was kept undisturbed at 28 $^\circ\text{C}$ about 5 hours after being stirred for 1 minute.

c. Purification of the AuNRs

A 10 mL AuNR solution was centrifuged at 8000 rpm for 30 minutes and then the supernatant was removed with a pipette. The pellet was suspended in 10 mL water. The solution was centrifuged at 3000 rpm for 25 minutes and the supernatant was collected. The AuNR solution was then centrifuged at 8000 rpm for 30 minutes. The supernatant was discarded. The pellet was suspended in 100 μL water. The concentration was measured by a UV spectrometer.

4. Preparation of the AuNRs modified with thiolated DNA

The disulfide bonds in the thiolated oligonucleotides were reduced to monothiol using TCEP (20mM, 1h) in water. The oligonucleotides were purified using size exclusion columns (G-25, GE Healthcare) to remove small molecules.

The purified DNA was added to the AuNR solution ($\text{OD} \sim 1$) containing 0.01% (w/v) SDS with a molecular ratio of 4000: 1. The mixture solution was incubated for 6 hours at 28 $^\circ\text{C}$. Then the buffer concentration was adjusted to 1 $\times$ TBE (89 mM Tris, 89 mM boric acid, 2 mM EDTA, pH 8.0) from a 10 $\times$ TBE buffer. After that, a 5M NaCl solution was slowly added until the final concentration of NaCl reached 500 mM in 24 hours.

Subsequently, the AuNR-DNA conjugates were centrifuged at 8000 rpm for 25 minutes. The pellet was suspended in a 1 mL 0.5 $\times$ TBE buffer containing 200 mM NaCl while the supernatant was discarded. The same centrifugation procedure was repeated three times to completely remove the excess thiolated DNA.

5. Self-assembly of the AuNRs on the DNA origami templates

The purified rectangular DNA origami structures were mixed with the AuNR-DNA conjugates at a ratio of two AuNRs for each binding group on the DNA origami. The mixture was annealed from 45 $^\circ\text{C}$ to 25 $^\circ\text{C}$ for 30 cycles in 2 hours.

6. Purification of the DNA origami assembled AuNRs

The annealed product of the DNA origami assembled AuNRs was loaded in a 0.8% Ethidium Bromide stained agarose gel (running buffer 1 $\times$ TAE/ Mg^{2+} , loading buffer 60% glycerol, 15 V/cm). Selected bands were cut out and the DNA origami assembled AuNRs were extracted from the gel with Freeze-Squeeze columns (Bio-Rad) at 4 $^\circ\text{C}$ or by electro-elution with dialysis tubing membranes (MWCO: 50K).

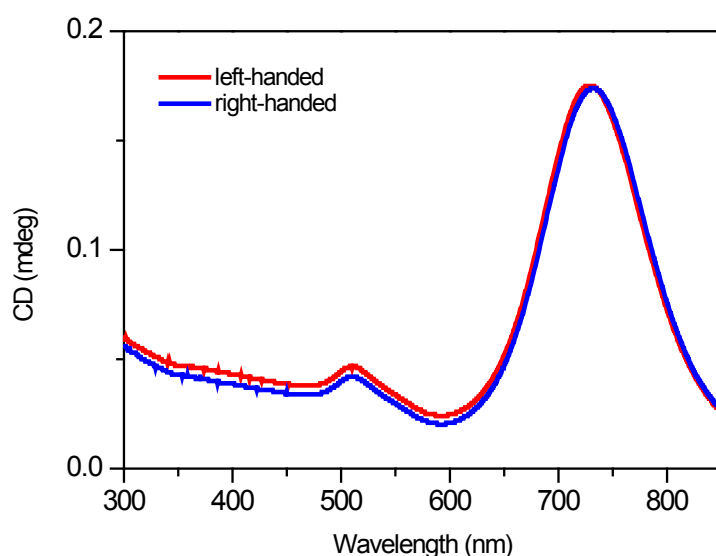


Fig. S1 Extinction spectra of the left- (red) and right-handed (blue) twisted AuNR structures.

7. TEM characterization of the DNA origami assembled AuNRs

The samples for TEM imaging were prepared by depositing a 4 μ L of the sample solution on a carbon-coated TEM grid. After 10 min, the excess solution was wicked from the grid using filter paper. To remove the deposited salt, the grid was washed with a droplet of water and the excess water was wicked away using filter paper. A droplet of a 0.7% uranyl acetate solution was used to treat the grid and the excess solution was wicked away. Then, the grid was treated with a second droplet of the uranyl acetate solution for 40 seconds and the excess solution was removed using filter paper. All the grids were kept at room temperature. TEM studies were conducted using a Tecnei G2-20S TWIN, operating at 200 kV in a dark field mode.

8. Circular Dichroism measurements

The CD spectra were measured by a Jasco J-815 spectropolarimeter using the following parameters: wavelength range, 200 nm to 900 nm; optical length, 0.5 cm; scanning speed, 200 nm/min.

Quantitative analysis of bands on the agarose gel was performed using Image J (<http://rsb.info.nih.gov/ij/>). The target bands yields of left-handed crosses and right-handed crosses were 65.1% and 67.4%, respectively. Target bands were cut out; the AuNRs crosses were extracted from the gel and imaged with TEM. The yields on the TEM images were counted to be 80.5% (91/113) for left-handed structures and 82.2% (88/107) for right-handed structures.

B. Supporting figures and legends

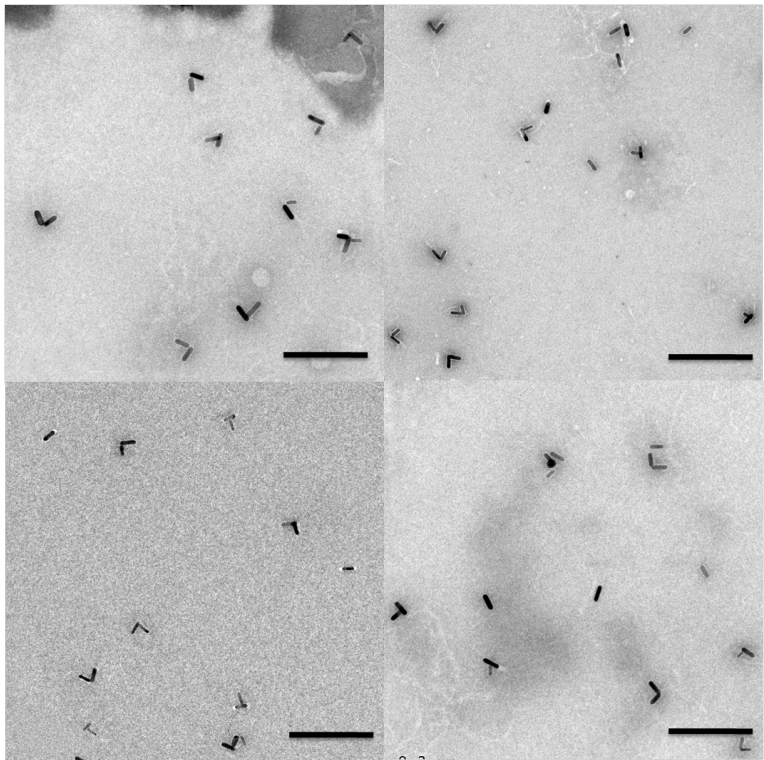
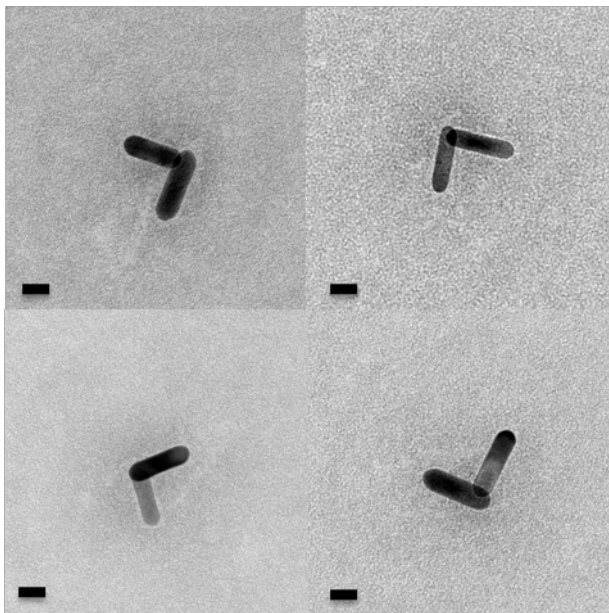


Fig. S2 Additional TEM images of the left-handed twisted AuNR structures (top scale bar: 20 nm; bottom scale bare: 200 nm).

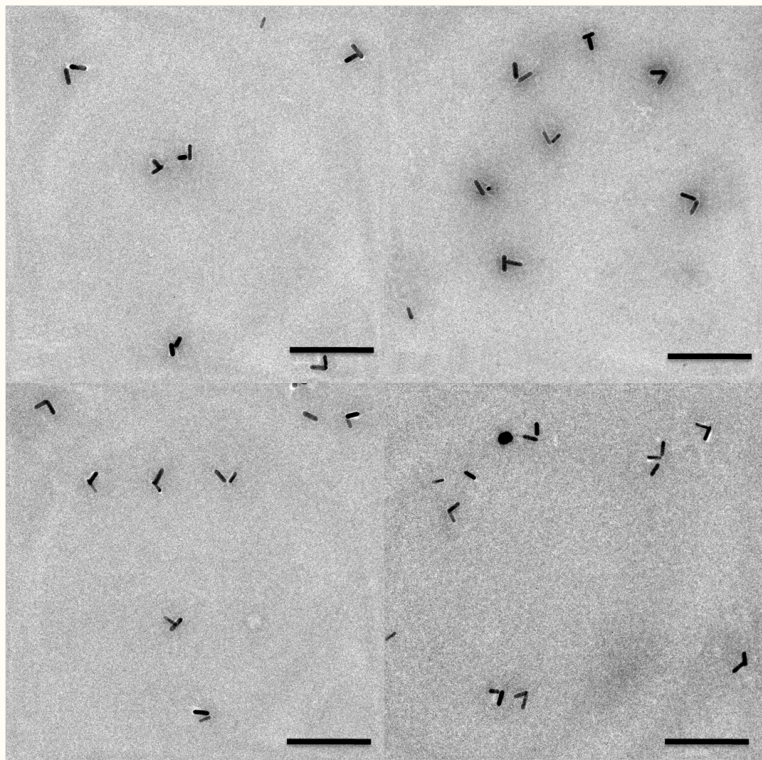
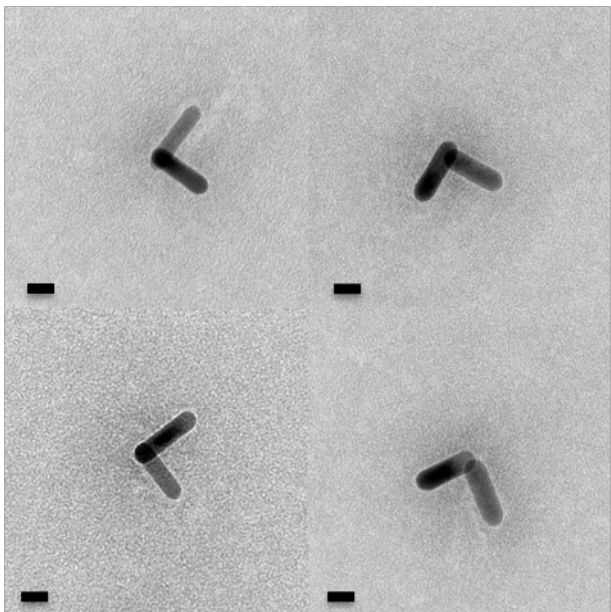


Fig. S3 Additional TEM images of the right-handed twisted AuNR structures (top scale bar: 20 nm; bottom scale bare: 200 nm).

C. DNA strands used in the experiment

1. Capture strands

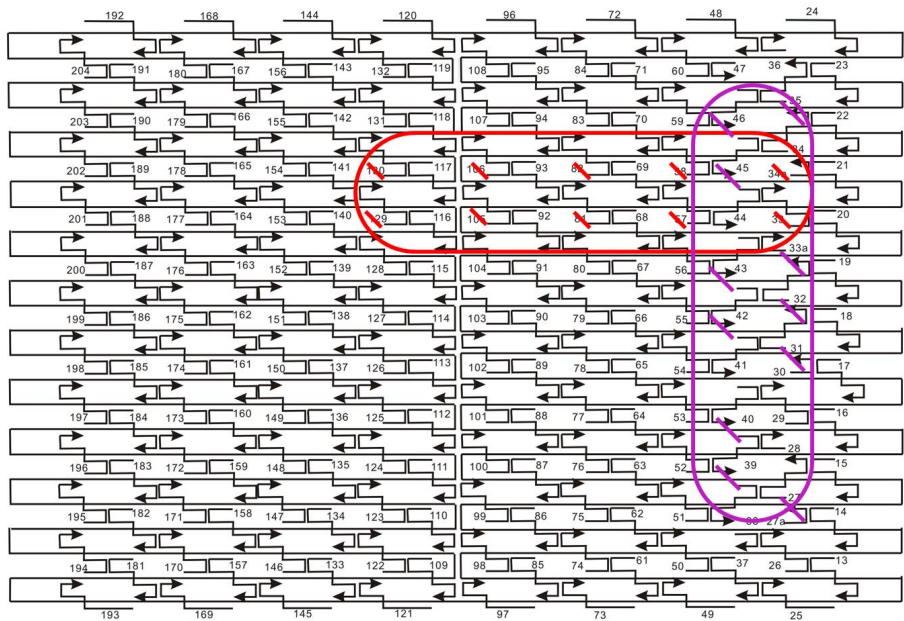


Fig. S4 Design of the left-handed twisted AuNR structure.

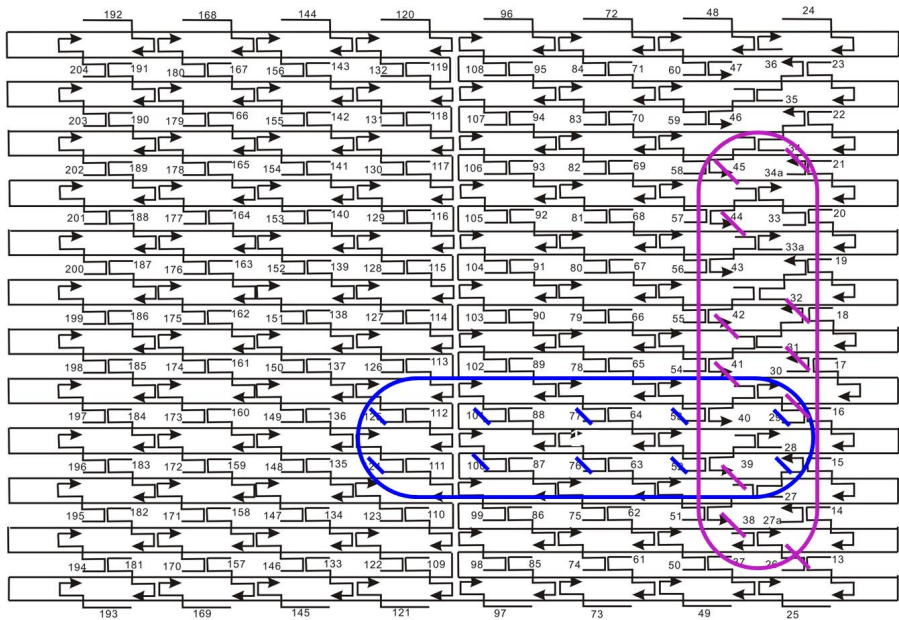


Fig. S5 Design of the right-handed twisted AuNR structure.

Binding Site (red)

R29, AAAAAAAAAAAAAAAAAA ACCCGTCGTCATATGTACCCCGGTAAAGGCTA
R30, AAAAAAAAAAAAAAAAAA TCAGGTCAC TTTTGCG
R53, AAAAAAAAAAAAAAAAAA CTTTCATCCCCAAAAACAGGAAGACCGGAGAG
R54, AAAAAAAAAAAAAAAAAA GG TAGCTAGGATAAAAATTTTAGTTAACATC
R77, AAAAAAAAAAAAAAAAAA AAATAATTTTAAATTGTAAACGTTGATATTCA
R78, AAAAAAAAAAAAAAAAAA ACCGTTCTAAATGCAATGCCTGAGAGGTGGCA
R101, AAAAAAAAAAAAAAAAAA AGCTCATTTTCGCATTAAATTTTGAGCTTAGA
R102, AAAAAAAAAAAAAAAAAA AAGACAGTCATTCAAAAGGGTGAGAAGCTATAT
R125, AAAAAAAAAAAAAAAAAA ATAGAATCCCTGAGAAGAGTCAATAGGAATCAT
R126, AAAAAAAAAAAAAAAAAA AAATTACTACAAATTCTTACCAGTAATCCCATC

Binding site (blue)

R33, AAAAAAAAAAAAAAAAAA ACTGGATAACGGAACAACATTATTACCTTATG
R34a, AAAAAAAAAAAAAAAAAA CGATTTTAGAGGACAG
R57, AAAAAAAAAAAAAAAAAA TTTGCCAGATCAGTTGAGATTTAGTGGTTTAA
R58, AAAAAAAAAAAAAAAAAA TTTCAACTATAGGCTGGCTGACCTTGATCAT
R81, AAAAAAAAAAAAAAAAAA CCAAAATATAATGCAGATACATAAACACCAGA
R82, AAAAAAAAAAAAAAAAAA ACGAGTAGTGACAAGAACCGGATATACCAAGC
R105, AAAAAAAAAAAAAAAAAA CATAACCCGAGGCATAGTAAGAGCTTTTTAAG
R106, AAAAAAAAAAAAAAAAAA GAATAAGGACGTAACAAAGCTGCTCTAAAACA
R129, AAAAAAAAAAAAAAAAAA GCAATAGCGCAGATAGCCGAACAATTCAACCG
R130, AAAAAAAAAAAAAAAAAA ATTGAGGGTAAAGGTGAATTATCAATCACCGG

Binding site (purple)

R27, AATAATAATAATAATAATT TCGACTCTGCAAGGCGATTAAAGTTCGCATCGT
R31, AATAATAATAATAATAATT TGGAGAAGCAGAATTAGCAAAATTAAAGTACGG
R32, AATAATAATAATAATAATT TGTCTGGAAGAGGTCATTTTTCGCAGAAAAC
R33a, AATAATAATAATAATAATT GAGAATGAATGTTTAG
R35, AATAATAATAATAATAATT TGAGGACTAGGGAGTTAAAGGCCGAAAGGAAC
R39, AATAATAATAATAATAATT GATGTGCTAGAGGATCCCCGGGTACTTTCCAG

R40, AATAATAATAATAAATTATGTGAGCATCTGCCAGTTTGAGGGAAAGGGG
R41, AATAATAATAATAAATT AGATCTACTGATAATCAGAAAAGCAACATTAA
R42, AATAATAATAATAAATT TCAAGGCAACTTTATTTCAACGCAATTTTGTAG
R43, AATAATAATAATAAATT TTTTGATAAGTTTCATTCCATATACATACAGG
R44, AATAATAATAATAAATT TATAGTAAACCATAAATCAAAAATCATTGCTCC
R45, AATAATAATAATAAATT TGTGAATTACAGGTAGAAAGATTCAGGGGGTA
R46, AATAATAATAATAAATT TCGAAATCTGTACAGACCAGGCGCTTAATCAT

2. Sequences used in the assembly of the DNA origami template

13 TGGTTTTTAACGTCAAAGGGCGAAGAACCATC
14 CTTGCATGCATTAATGAATCGGCCCGCCAGGG
15 TAGATGGGGGGTAACGCCAGGGTTGTGCCAAG
16 CATGTCAAGATTCTCCGTGGGAACCGTTGGTG
17 CTGTAATATTGCCTGAGAGTCTGGAAAAGTAG
18 TGCAACTAAGCAATAAAGCCTCAGTTATGACC
19 AAACAGTTGATGGCTTAGAGCTTATTTAAATA
20 ACGAACTAGCGTCCAATACTGCGGAATGCTTT
21 CTTTGAAAAGAACTGGCTCATTATTTAATAAA
22 ACGGCTACTTACTTAGCCGGAACGCTGACCAA
23 GAGAATAGCTTTTGCGGGATCGTCGGGTAGCA
24 ACGTTAGTAAATGAATTTTCTGTAAGCGGAGT
25 ACCCAAATCAAGTTTTTTGGGGTCAAAGAACG
26 TGGACTCCCTTTTCACCAGTGAGACCTGTCGT
27a GCCAGCTGCCTGCAGG
27 TCGACTCTGCAAGGCGATTAAGTTCGCATCGT
28 AACCGTGCGAGTAACA
29 ACCCGTCGTCATATGTACCCCGGTAAAGGCTA
30 TCAGGTCACTTTTGCG
31 GGAGAAGCAGAATTAGCAAAATTAAAGTACGG
32 TGTCTGGAAGAGGTCATTTTTGCGCAGAAAAC

33a GAGAATGAATGTTTAG
33 ACTGGATAACGGAACAACATTATTACCTTATG
34a, CGATTTTAGAGGACAG
34 ATGAACGGCGCGACCT GCTCCATGAGAGGCTT
35 TGAGGACTAGGGAGTTAAAGGCCGAAAGGAAC
36 AACTAAAGCTTTCCAG
37 AGCTGATTACAAGAGTCCACTATTGAGGTGCC
38 CGGGAAACGGGCAAC
39 GATGTGCTAGAGGATCCCCGGGTACTTTCCAG
40 ATGTGAGCATCTGCCAGTTTGAGGGAAAGGGG
41 AGATCTACTGATAATCAGAAAAGCAACATTAA
42 TCAAGGCAACTTTATTTCAACGCAATTTTGTAG
43 TTTTGATAAGTTTCATTCCATATACATACAGG
44 TATAGTAAACCATAAATCAAAAATCATTGCTCC
45 TGTGAATTACAGGTAGAAAGATTCAGGGGGTA
46 TCGAAATCTGTACAGACCAGGCGCTTAATCAT
47, AGGCTTGCAAAGACTTTTTCATGAAAATTGTG
47a, AGGCTTGCAAAGACTT
48 CGTAACGATCTAAAGTTTTGTCGTGAATTGCG
49 GTAAAGCACTAAATCGGAACCCTAGTTGTTCC
50 AGTTTGGAGCCCTTCACCGCCTGGTTGCGCTC
51 ACTGCCCCGCCGAGCTCGAATTCGTTATTACGC
52 CAGCTGGCGGACGACGACAGTATCGTAGCCAG
53 CTTTCATCCCCAAAAACAGGAAGACCGGAGAG
54 GGTAGCTAGGATAAAAATTTTAGTTAACATC
55 CAATAAATACAGTTGATTCCCAATTTAGAGAG
56 TACCTTTAAGGTCTTTACCCTGACAAAGAAGT
57 TTTGCCAGATCAGTTGAGATTTAGTGGTTTAA
58 TTTCAACTATAGGCTGGCTGACCTTGTATCAT

59 CGCCTGATGGAAGTTTCCATTAAACATAACCG
60 ATATATTCTTTTTTCACGTTGAAAATAGTTAG
61 GAGTTGCACGAGATAGGGTTGAGTAAGGGAGC
62 TCATAGCTACTCACATTAATTGCGCCCTGAGA
63 GAAGATCGGTGCGGGCCTCTTCGCAATCATGG
64 GCAAATATCGCGTCTGGCCTTCCTGGCCTCAG
65 TATATTTTAGCTGATAAATTAATGTTGTATAA
66 CGAGTAGAACTAATAGTAGTAGCAAACCCTCA
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85 CCAGCAGGGGCAAAATCCCTTATAAAGCCGGC
86 GCTCACAATGTAAAGCCTGGGGTGGGTTTGCC
87 GCTTCTGGTCAGGCTGCGCAACTGTGTTATCC
88 GTTAAAATTTTAACCAATAGGAACCCGGCACC

89 AGGTAAAGAAATCACCATCAATATAATATTTT
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202 ACGCAAAGGTCACCAATGAAACCAATCAAGTT
203 TGCCTTTAGTCAGACGATTGGCCTGCCAGAAT
204 GGAAAGCGACCAGGCGGATAAGTGAATAGGTG

References

1. Kyle Lund et. al. Nature 2010, **465**, 206-310.
2. B. Nikoobakht and M. A. El-Sayed, Chem. Mater. 2003, **15**, 1957.

